

First results from the CRIS experiment

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Abstract The ability to study rare isotopes with techniques such as mass spectrometry and laser spectroscopy is often prevented by low production rates and large isobaric contamination. This has necessitated the development of novel beam cleaning techniques that can efficiently isolate the isotope of interest. The Collinear Resonance Ionization Spectroscopy (CRIS) experiment at ISOLDE, achieves this by resonantly ionizing a bunched atom beam in a region of ultra high vacuum. This method is motivated by the need to measure the hyperfine structure and isotope shift at the extremes of isospin where typical production rates drop to 1 atom/s. The technique also offers the ability to purify an ion beam and even select long-lived isomeric states (>1 ms) from the ground state, which can be subsequently studied by decay spectroscopy or mass spectrometry experiments. This paper will report on the successful commissioning of the CRIS beam line and the recent laser spectroscopy results and laser assisted nuclear decay spectroscopy on the neutron deficient francium isotopes.

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1 Introduction

Optical studies of radioactive nuclei provide a wealth of model-independent nuclear observables: changes in mean-square charge radii, nuclear moments and spin. These observables provide critically important information on the composition of the nuclear wave function, the distribution of charge and shape of the nucleus. It should also be noted that the simple measurement of additional resonances within the hyperfine structure can unambiguously demonstrate the existence of long-lived isomeric states [1]. If the status of measurements are plotted on the nuclear chart, gaps in the knowledge are apparent. These gaps arise due to production difficulties at radioactive beam facilities or an absence of suitable transitions for laser spectroscopy. The limits of known nuclear existence are beyond the limits of optical measurements for all but the lightest elements, which reflects the relatively low sensitivity of existing techniques [2]. There are two main families of techniques that are currently used at on-line facilities: collinear and in-source laser spectroscopy. Collinear laser spectroscopy was first experimentally demonstrated by W.E Lamb's group [3]. They showed that by collinearly overlapping the laser and electrostatically accelerated ion or atom beam the Doppler broadening associated with thermal motion at the source is compressed due to conservation of energy, with an observed linewidth ranging from 7–25 MHz. The collinear beams method is versatile and has been adapted in a variety of ways to take advantage of the properties of specific elements [4]. With a few exceptions these variations can study radioactive species produced with yields down to 1000 atoms/s [2]. In-source laser spectroscopy has been developed to reach the highest sensitivity and utilizes the laser ion source production method [5–7] to measure the hyperfine structure and isotope shift. This method has been able to study cases with production rates of less than 1 atom per second [8, 9] but with a resolution much lower than is possible in the collinear geometry (3–4 GHz). When the method is utilized in a supersonic gas jet it is possible to reduce this linewidth by more than an order of magnitude (~ 200 MHz) [10].

The combination of collinear and resonance ionization spectroscopy (CRIS) was first proposed by Yu. A. Kudriavtsev and V. S. Letokhov in 1982 as a novel method for detecting rare isotopes with natural abundances below 10^{-10} [11]. The method aims at combining the intrinsic high resolution of the collinear geometry with the high efficiency of resonance ionization and charge particle detection. The application of the method at on-line facilities such as ISOLDE was proposed by V. S. Letokhov in collaboration with members of the IS80 Experiment at ISOLDE [12]. The results from the first successful experiment were reported in 1988 [13]. A parallel programme utilized the method for trace analysis of ^{40}K , ^3He , $^{89,90}\text{Sr}$ in Troitzk and Mainz [14–16]. The ability of the CRIS technique to select a particular isotope from a heavily contaminated beam was demonstrated in the initial experiments [17]. A signal-to-noise ratio of better than 20 was observed for the neutron deficient isotopes of ytterbium. The first experiments successfully measured $^{157,159}\text{Yb}$ with a linewidth of 50 MHz and a high ionization efficiency of approximately 80 % [17]. However the total experimental efficiency was 0.001 % due to the low population of the metastable state through charge exchange and the losses associated with the duty cycle of the laser and a continuous ion beam. The need for a pulsed laser ion source to avoid these losses was highlighted in the original proposal [5, 12].

The introduction of the radio-frequency linear Paul trap at the IGISOL facility [18–20] in 2000 presented an opportunity to test the CRIS method with bunched beams [21]. A

demonstration experiment was performed with ^{27}Al using the $^2P_{1/2} - ^2D_{3/2}$ (308 nm) transition for the resonant step and 532 nm to non-resonantly ionize the bunch. The resonant light was produced by frequency doubling laser light from a pulse dye amplified system. A total experimental efficiency of 3 % was determined, which represents a factor of more than 1000 improvement compared to the previous CRIS experiments with continuous beams. This demonstration experiment also highlighted the need for ultra-high vacuum (UHV) in the interaction region to avoid collisional ionization of the fast atomic beam. At a pressure of 10^{-5} mbar the non-resonant ionization efficiency reaches 1 %.

A programme of bunched-beam spectroscopy at ISOLDE was proposed by the Manchester group in 2003 [22]. This supported the installation of an radio-frequency linear Paul-trap (ISCOOL) after the ISOLDE high resolution separator (HRS) [23]. This device has now greatly extended the reach of the collinear laser spectroscopy programme at ISOLDE, allowing measurements of short-lived nuclei produced with rates down to 10^3 atoms/s [24–27]. The ability to cool and bunch beams made it possible to further develop the CRIS technique and continue where the original work has stopped at ISOLDE.

2 Recent development of CRIS at ISOLDE

In 2008 a new proposal to establish a dedicated CRIS experiment at ISOLDE was submitted [28]. The decommissioning of the COMPLIS experiment [29] provided space in the hall to begin installation of the apparatus for the CRIS beam-line and pulsed lasers. During the period of 2008–2009 the main parts of the CRIS experiment were designed and installed into the ISOLDE hall. A technical drawing of the experimental beam line is shown in Fig. 1. The CRIS apparatus can take beam from either ISOLDE or an auxiliary ion source located before the initial quadrupole triplet. A 34° electrostatic bend is used to collinearly overlap the ion beam with the laser beams. An aperture has been made into one of the bending plates in the 34° bend to allow the laser beam to be launched into the beam line. Simulations were performed with SIMION to verify that this slot introduces a negligible perturbation to the ion beam's path. A charge exchange cell is located after the 34° bend, which is typically loaded with potassium to neutralize ion beams of francium [30]. A section of differential pumping separates the high-vacuum ($< 10^{-5}$ mbar) region associated with the charge exchange cell from the interaction region which is kept at UHV ($< 10^{-8}$ mbar). A single plate is used to electrostatically deflect the non-neutralized component within the differential pumping region. The interaction region has a length of 1.2 m, which equates to a $3.5 \mu\text{s}$ transit time for a 30 keV beam of $A = 50$, well matched to the bunch length of $3 \mu\text{s}$ produced by the ISCOOL [31]. After the interaction region a 20° electrostatic bend is used to deliver the resonantly ionized bunch to the detection chamber. The CRIS experiment utilizes charge particle detection, with a microchannel plate (MCP) and passivated implanted planar silicon (PIPS) detectors [32].

3 On-line commissioning of the CRIS experiment

Measurements of short-lived francium isotopes were carried out to commission the CRIS experiment. The radioactive isotopes were produced by impinging 1.4 GeV protons onto a uranium carbide target. The target was heated to $\sim 2000^\circ\text{C}$, which allowed a rapid evacuation of the short-lived species. The atoms were surface ionized, accelerated to 50 keV and mass separated. The ion beam was then cooled and bunched using the ISCOOL

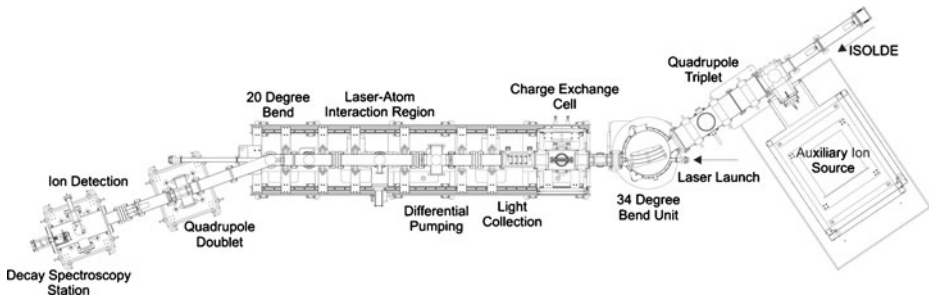


Fig. 1 Technical drawing of the CRIS beam line

device. A bunch width of $3 \mu\text{s}$ was measured for the francium isotopes corresponding to 0.5 m spatial dimension [33]. The trapping time was set to match the 30 Hz pulse repetition rate of the ionization laser. The bunched beam was delivered to the CRIS beam line where it was neutralized and overlapped with two pulsed lasers. The experiment studied $^{202-206,218,219,229,231}\text{Fr}$ using a two-step ionization scheme, which consisted of a resonant step from the $7s^2S_{1/2}$ ground state to the $8p^2P_{3/2}$ excited state ($23\,658.306\text{ cm}^{-1}$ [34]) and a non-resonant excitation to the continuum using the fundamental frequency from an Nd:YAG laser. The resonant step was excited with light from a narrow-bandwidth pulsed (10 kHz) Ti:Sapphire laser that was frequency doubled and fiber coupled from the RILIS cabin [35]. A trigger from the 10-kHz Nd:YAG pump laser was used to synchronize the ionizing Nd:YAG laser, ISCOOL, and the data acquisition system. The frequency doubled light had a measured linewidth of 1.5 GHz, sufficient for extracting the magnetic moments and charge radii of the francium isotopes [36]. Up to 120 mW of 422-nm light was transported through the multi-mode fiber to the CRIS experiment. In the initial experiment a maximum of 15 mJ/pulse of 1064 nm was transported through the interaction region of the CRIS beam line. The excitation frequency of the resonant transition was scanned in this experiment rather than the velocity of the atomic bunch. A HighFinesse WS7 wavelength meter was used to continuously measure the frequency during this experiment. The resonantly ionized isotopes of francium were transported to either the MCP detector or the decay spectroscopy station [32].

Examples of typical spectra obtained with the CRIS method are shown in Fig. 2, which presents the neutron deficient odd-A francium isotopes. The data has been presented with a logarithmic scale so that the background rate is visible. The background consists of collisionally ionized atoms, activation of the MCP assembly and the dark count rate of the MCP. The initial activation occurred with ^{221}Fr , which was used for beam transport and neutralization efficiency measurements. While ^{221}Fr has a 4.9 m half-life the progeny isotopes ^{213}Bi and ^{209}Pb have half-lives of 45.6 m and 3.25 h respectively, resulting in an instantaneous activity of greater than 10 kBq. This was reduced by a factor of at least 10^4 by gating the detected counts with the arrival time and the temporal length of the ion bunch (FWHM $\sim 2 \mu\text{s}$). Even with such a high rejection the observed background in Fig. 2 is dominated by the activation of the plates. This initial activation was allowed to decrease before the cases with the lowest yields were attempted such as ^{202}Fr . In order not to further activate the MCP assembly when reference scans of ^{221}Fr were made only a very small fraction of the beam was delivered. This also avoided any systematic effects associated with saturating the ISCOOL. For the majority of accessible elements at ISOLDE a stable reference isotope is utilized, which will not introduce such activation problems in the future.

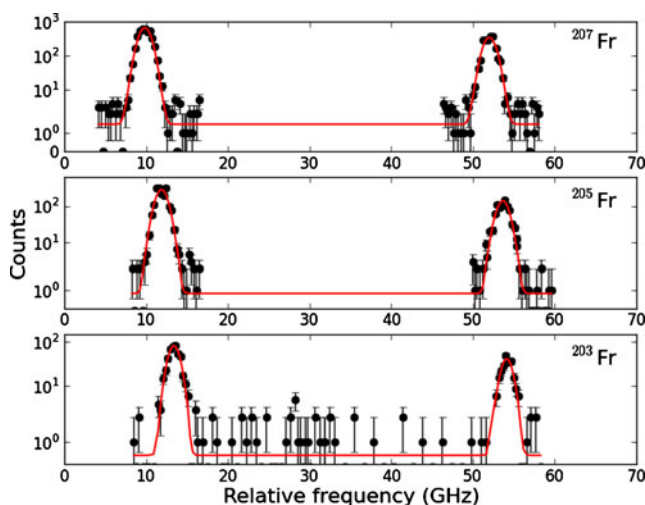


Fig. 2 Hyperfine structure spectra of $^{203,205,207}\text{Fr}$ measured with the CRIS technique

From the measured yields of ^{202}Fr and ^{218}Fr it was possible to determine a total experimental efficiency of 1 %. This represents a factor of 1000 increase compared to the first on-line CRIS measurements of ytterbium [17]. This method is a factor of 100 more sensitive than the state-of-the-art fluorescence detection techniques used to measure $^{204,205,206}\text{Fr}$ [37]. The increased sensitivity of the CRIS technique is in part due to the high experimental efficiency but also the high background rejection. During the ^{202}Fr measurement a background rate of $2 \times 10^{-3} \text{ s}^{-1}$ was determined. This sets an upper limit for the non-resonant ionization efficiency as 0.001 % based on the yield of the ^{202}Tl isobar [36].

Regular reference scans were taken of ^{221}Fr that measured the hyperfine structure of the $^2S_{1/2}$ state. From these measurements a linewidth of 1.5 GHz was determined for the second harmonic of the narrow bandwidth Ti:Sapphire laser [35]. The linewidth of the laser was continuously measured with the wavelength meter to avoid multimode lasing. The reference hyperfine structure measurements were used to study the long- and short-term frequency stability of the experiment. A 30 MHz scatter in the measured $A(^2S_{1/2})$ factor of ^{221}Fr was determined from the reference scans [38]. This was used to estimate the minimum error of new measurements. There was a short term fluctuation of ± 100 MHz with a long term drift of ~ 180 MHz in the centre of gravity of ^{221}Fr . This was taken into account in the estimation of the experimental error in the measured isotope shifts. This work highlights the suitability of the narrow-bandwidth pulsed Ti:Sapphire laser for in-source laser spectroscopy [39].

4 Summary

The CRIS beam line and laser laboratory has been completed and commissioned. The first on-line demonstration of the technique has been carried out by studying the radioactive element francium. This work has demonstrated a total experimental efficiency of 1 % based on the yields of $^{202,218}\text{Fr}$. New hyperfine structure measurements have been made on nine isotopes produced with yields down to 100 atoms per second. The process of non-resonant ionization is suppressed by the experimental apparatus by more than a factor of 10^5 . This

demonstrates the suitability of the CRIS technique for isobar cleaning and isomer selection. A parallel research programme has investigated the ability to purify radioactive beams and select just the isomeric states [40, 41]. The hyperfine structure of the isomers in ^{204}Fr have been measured and the selected nuclear states directed to the decay spectroscopy station and studied. This work is the subject of a forth coming paper [42]. The CRIS collaboration is now investigating increasing the resolution of the experiment through the use of pulse amplification and injection seeded techniques. A resonance linewidth of 50 MHz has been previously demonstrated [17]. Proposals to study the neutron rich copper and gallium isotopes have been accepted. Further studies of the isomers in $^{201,203}\text{Fr}$ are also planned.

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